Argonne
NATIONAL LABORATORY

ANL-AFCI-179

Effect of Reprocessing and Recycling on the Geologic Repository Dose Rate: Status

Nuclear Engineering Division

About Argonne National Laboratory

Argonne is a U.S. Department of Energy laboratory managed by UChicago Argonne, LLC under contract DE-AC02-06CH11357. The Laboratory's main facility is outside Chicago, at 9700 South Cass Avenue, Argonne, Illinois 60439. For information about Argonne, see www.anl.gov.

Availability of This Report

This report is available, at no cost, at http://www.osti.gov/bridge. It is also available on paper to the U.S. Department of Energy and its contractors, for a processing fee, from:

U.S. Department of Energy
Office of Scientific and Technical Information
P.O. Box 62
Oak Ridge, TN 37831-0062
phone (865) 576-8401
fax (865) 576-5728
reports@adonis.osti.gov

ANL-AFCI-179

Effect of Reprocessing and Recycling on the Geologic Repository Dose Rate: Status

by
E.E. Morris, W.M. Nutt, and R.A. Wigeland
Nuclear Engineering Division, Argonne National Laboratory

September 28, 2006

Effect of Reprocessing and Recycling on the Geologic Repository Dose Rate: Status

By
E. E. Morris, W. M. Nutt, and R. A. Wigeland
Nuclear Engineering Division
Argonne National Laboratory, Argonne, Illinois

Introduction

Two simplified repository performance assessment models are used to assess the impact of modeling changes in on conclusions regarding the impact of various reprocessing and recycling strategies. Waste streams from a pressurized water reactor (PWR) and a preliminary design for an advanced burner test reactor (ABTR) are used for this study of the effects on the estimated dose rate resulting from the release of radionuclides from a geologic repository. Calculations for the PWR make use of radionuclide discharge vectors for an assumed burnup of 51 GWd/MTIHM[1]. The repository is assumed to be filled with 70,000 MT of the spent fuel or with a glass waste form containing the radionuclides from 70,000 MT of spent PWR fuel. For the ABTR, the radionuclide inventory discharged at the end of an equilibrium cycle[2] is processed into a glass waste form for repository disposal, assuming actinide recovery efficiencies ranging from 90% to 99.99%. The recovered actinides are returned to the reactor. To compare with the PWR results, the repository is assumed to be filled with ABTR waste from fuel that has generated the same amount of thermal energy as 70,000 MT of the PWR fuel.

The two repository performance assessment models, the first a simplified model[3] (SSR) based on the site recommendation model used by the Yucca Mountain Project (YMP)[4], and the second an updated simplified model (US) based on more recent modeling developments by the YMP are implemented in the computer simulation code GoldSim[5]. The updated model is based on a simplified model used to conduct a sensitivity analysis to evaluate factors that potentially influence performance of a repository at Yucca Mountain over the period of peak dose[6]. Factors that have either a minor or no effect on the peak dose either were not included in that simplified model or were included in a bounding representation. In the US model, enhancements were made to include some factors that have an effect on the dose occurring earlier in time following repository closure (e.g., within several tens of thousands of years). These enhancements are in the form of simplified sub-models that include:

- early waste package failure;
- stress corrosion cracking of the waste package outer barrier; and
- waste form degradation
    - commercial spent nuclear fuel cladding
    - commercial spent nuclear fuel (CSNF) waste form degradation
    - defense high level waste (DHLW) waste form degradation
    - diffusive radionuclide transport through the EBS

Comparison Calculations

Calculations for a repository filled with 70,000 MT of spent PWR fuel, directly disposed, are shown in Figs. 1 and 2. The results in Fig. 1 were evaluated using the SSR model while

those in Fig. 2 were obtained using the US model. All results are normalized to the peak mean dose rate as determined using the SSR model. In both figures, in addition to showing the total dose rate, the dose-rate contributions from several individual radionuclides are also shown. The most striking difference between the calculations using the SSR and the US models is the fact that most waste packages do not fail until about 500,000 years in the newer model. Waste package corrosion rates in the SSR model did not vary with temperature and were held constant at the 60°C rates even though the repository cools with time. Waste package corrosion rates in the US model vary with temperature and decrease as the repository temperature decreases. This results in longer waste package lifetimes. A second difference between the two calculations is that the radionuclides contributing most to the total dose rate are different. In the SSR model, the principle contributors to the dose rate are ^{237}Np, ^{226}Ra, ^{230}Th, and ^{210}Pb. With the US model ^{237}Np is still one of the major contributors, but ^{242}Pu is the largest contributor to the mean dose rate and ^{233}U in the third most important contributor. ^{230}Th is a much less important contributor to the dose rate in the US model while ^{234}U, ^{236}U, and ^{238}U are much more important contributors than in the SSR model. While ^{226}Ra and ^{210}Pb are not modeled in the US model, since these isotopes are shorter lived progeny of the ^{230}Th, their dose rate contributions would be expected to be similar to that of ^{230}Th and, therefore, relatively unimportant. These shifts in isotopic importance are primarily the result of using updated biosphere dose conversion factors in the US model. The updated factors are based on the most recent recommendations of the International Commission on Radiation Protection.

Since for the ABTR calculations, the waste will be assumed to be immobilized in glass, the PWR calculations described in the foregoing paragraph were repeated with the same radionuclide inventory but with the waste immobilized in glass rather than directly disposed as spent fuel. The results from calculations for the glass waste form are compared in Fig. 3 with the direct disposal calculations from the preceding paragraph. They show that the glass waste form used in the US model results in a reduction in the total mean dose rate whereas in the SSR model, the use of the glass waste form results in an increased dose rate. The glass waste-form sub-model used in the US model produces lower degradation rates than the model used in the SSR model.

Several performance assessment calculations using both the SSR and the US models were carried out for the waste streams from ABTR equilibrium-recycle cores with low, medium, and high actinide conversion ratios. As noted in the introduction, the calculations assume that ABTR waste placed in repository was from spent fuel that had produced the same amount of thermal energy as 70,000 MTIHM of spent PWR fuel with 51 GWd/MTIHM of burnup. Several cases were considered for the ABTR fuel ranging from disposal of the entire radionuclide inventory in a glass waste form to disposal in a glass waste form with 90%, 99%, 99.9%, and 99.99% of the actinides removed. All dose rate results at a well located approximately 20 km from the repository were normalized to the peak mean dose rate calculated by the SSR model for the case of direct disposal of 70,000 MTIHM of spent PWR fuel. Results from the calculations with the SSR model are summarized in Table 1. Also included in the table are results for the case where the entire PWR radionuclide inventory is immobilized in a glass waste form. In the table, the normalized peak mean dose rates, the normalized mean peak dose rates, and the normalized median peak dose rates for the three ABTR conversion ratio cases are compared with the corresponding dose rates for the spent PWR fuel cases.

The results in Table 1 show that the lower the actinide removal efficiency, the greater the advantage of a higher conversion ratio with respect to dose reduction for a given energy generation. If an actinide removal efficiency of 99.99% can be achieved, then the differences between high and low conversion ratio dose reductions are much smaller. The dose reductions shown in Table 1 indicate the potential for increased efficiency in repository utilization. Thermal analysis and more detailed waste form definition will be required to determine the form and magnitude of the efficiencies that can actually be achieved.

In Table 2, lists results using the US model to repeat the set of calculations described in the foregoing paragraph. As before, dose rates from the waste streams for various ABTR conversion ratios were compared with the dose rate resulting from the direct disposal of spent nuclear fuel from a PWR with 51 GWd/MTIHM burnup. Note that the results in Table 2 are normalized by the peak mean dose rate for the direct disposal of the spent PWR fuel as calculated with the US model. Just as in the previous cases, the thermal energy produced by the fuel from which the waste streams result is the same as that produced by the PWR fuel. Also as before, the calculations assume that the ABTR waste will be immobilized in borosilicate glass. Because, as noted earlier, the US glass waste form is more effective than spent fuel in retaining radionuclides, all glass waste form results in Table 2 are lower than the corresponding results in Table 1. This is the case even though the normalizing factor is smaller for the results in Table 2. Both the US calculations and the SSR calculations show that the lower the actinide removal efficiency, the more advantageous is the higher conversion ratio core.

Conclusions

Calculations for PWR spent fuel show that the updated simplified model produces a lower peak mean dose rate than the simplified site recommendation model used in previous analyses. Both models indicate that the peak dose rate occurs before 1,000,000 years, but the peak occurs at around 760,000 years in the updated model. In the simplified site recommendation model, the peak dose rate occurred at about 350,000 years. The difference in the time of the peak dose rate is caused by the fact that in the simplified site recommendation model, most waste packages have failed by 100,000 to 200,000 years whereas in the updated model very few packages fail until nearly 500,000 years. Further, while ^{237}Np is one of the most important contributors over the period of the highest dose rates for both models, different radionuclides make up the remaining important contributors. For example, ^{242}Pu was the fifth most important contributor to the dose rate in the simplified site recommendation model but is the most important contributor in the updated model. The reduced dose rate indicated by the updated model and the shift in the important isotopic contributors to the dose rate is caused by the use of biosphere dose conversion factors based on the most recent recommendations of the International Commission on Radiation Protection and, to some extent, changes in the solubilities and the partition coefficients for the actinide elements. Peak dose-rate contributions from the highly soluble and non-sorbing elements iodine and technetium agree within a factor of two or three between the two models.

If the spent PWR fuel was processed and all the radionuclides immobilized in borosilicate glass, the simplified site recommendation model indicates that the peak dose rate would increase, but less than a factor of two. The updated simplified model indicates that with

the entire radionuclide immobilized in glass, the peak dose rate would decrease by as much as a factor of five.

In spite of the differences between the results predicted by the two models, when the impact of actinide removal from the waste stream of the ABTR is considered, the models produce important agreement. As calculated with the updated simplified model, the peak dose rates when various fractions of the actinide content are removed is significantly smaller relative to that for PWR fuel than in the same type of calculation performed with the simplified site recommendation model. This is because of the improved performance in the updated model of the glass waste form relative to the performance of directly disposed spent fuel. However, both models agree that from the prospective of the repository dose rate, increased efficiency in the recovery of actinides for recycle in the ABTR reduces the importance of the conversion ratio achieved in the ABTR design. Stated another way, the higher the conversion ratio, the less sensitive the dose rate is to the actinide recovery efficiency.

References

1. J.A. Stillman, Argonne National Laboratory, Personal Communication, November 2004.

2. T. K. Kim, Argonne National Laboratory, Personal Communication, June 2006.

3. Eric Zwahlen, Golder Associates, Personal Communication, May 2002.

4. “Total System Performance Assessment for Site Recommendation,” TDR-WIS-PA-000001 REV 00 ICN 01 (December 2000).

5. “Users Guide: GoldSim Probabilistic Simulation Environment,” GoldSim Technology Group (January 2006).

6. Management and Technical Support Peak Dose Sensitivity Analysis, Prepared for the U.S. Department of Energy, November 2005. Available as part of the revisions to 40 CFR 197 at www.regulations.gov, EPA docket EPA-HQ-OAR-2005-0083, Document No. EPA-HQ-OAR-2005-0083-0352.

Table 1
Comparison of Normalized Dose Rates* for an ABTR with the Corresponding Dose Rates from Spent PWR Fuel. Results Calculated with the SSR Model.

	Direct Disposal of Spent PWR Fuel	Disposal of Spent PWR Fuel in Glass	ABTR No Actinide Removal	ABTR 90% Actinide Removal	ABTR 99% Actinide Removal	ABTR 99.9% Actinide Removal	ABTR 99.99% Actinide Removal
Low Conversions Ratio							
Median Peak	1.0663	0.5487	6.4758	1.1516	0.1887	0.0347	0.0104
Mean Peak	2.1653	1.9260	22.4487	4.2714	0.5151	0.0731	0.0207
Peak Mean	1.0000	1.4353	19.7661	3.2179	0.3510	0.0456	0.0132
Median Conversion Ratio							
Median Peak	1.0663	0.5487	2.4161	0.3981	0.0744	0.0163	0.0079
Mean Peak	2.1653	1.9260	9.3077	1.3025	0.1872	0.0345	0.0180
Peak Mean	1.0000	1.4353	7.4265	0.9247	0.1168	0.0208	0.0126
High Conversion Ratio							
Median Peak	1.0663	0.5487	1.1809	0.2150	0.0470	0.0133	0.0076
Mean Peak	2.1653	1.9260	4.7857	0.7625	0.1393	0.0317	0.0179
Peak Mean	1.0000	1.4353	3.2090	0.4512	0.0799	0.0191	0.0126

* Peak mean indicates the peak value of the time dependent mean dose rate. Mean peak refers to average value of the peak dose rate from each realization while the median peak is the median value of the peak dose rate from each realization. All calculations include 1000 realizations.

Table 2
Comparison of Normalized Dose Rates* for an ABTR with the Corresponding Dose Rates from Spent PWR Fuel. Results Calculated with the US Model.

	Direct Disposal of Spent PWR Fuel	Disposal of Spent PWR Fuel in Glass	ABTR No Actinide Removal	ABTR 90% Actinide Removal	ABTR 99% Actinide Removal	ABTR 99.9% Actinide Removal	ABTR 99.99% Actinide Removal
Low Conversions Ratio							
Median Peak	2.72251	0.12829	2.23306	0.28215	0.03155	0.00515	0.00240
Mean Peak	4.41738	0.37234	5.41397	0.78252	0.09156	0.01504	0.00939
Peak Mean	1.00000	0.18954	3.27943	0.40785	0.04392	0.00691	0.00337
Median Conversion Ratio							
Median Peak	2.72251	0.12829	0.64696	0.07227	0.00926	0.00279	0.00217
Mean Peak	4.41738	0.37234	1.72967	0.20990	0.02598	0.01015	0.00904
Peak Mean	1.00000	0.18954	0.92137	0.10215	0.01242	0.00391	0.00307
High Conversion Ratio							
Median Peak	2.72251	0.12829	0.24104	0.02594	0.00455	0.00230	0.00213
Mean Peak	4.41738	0.37234	0.70248	0.08159	0.01399	0.00932	0.00898
Peak Mean	1.00000	0.18954	0.34855	0.03636	0.00614	0.00329	0.00301

* Peak mean indicates the peak value of the time dependent mean dose rate. Mean peak refers to average value of the peak dose rate from each realization while the median peak is the median value of the peak dose rate from each realization. All calculations include 1000 realizations.

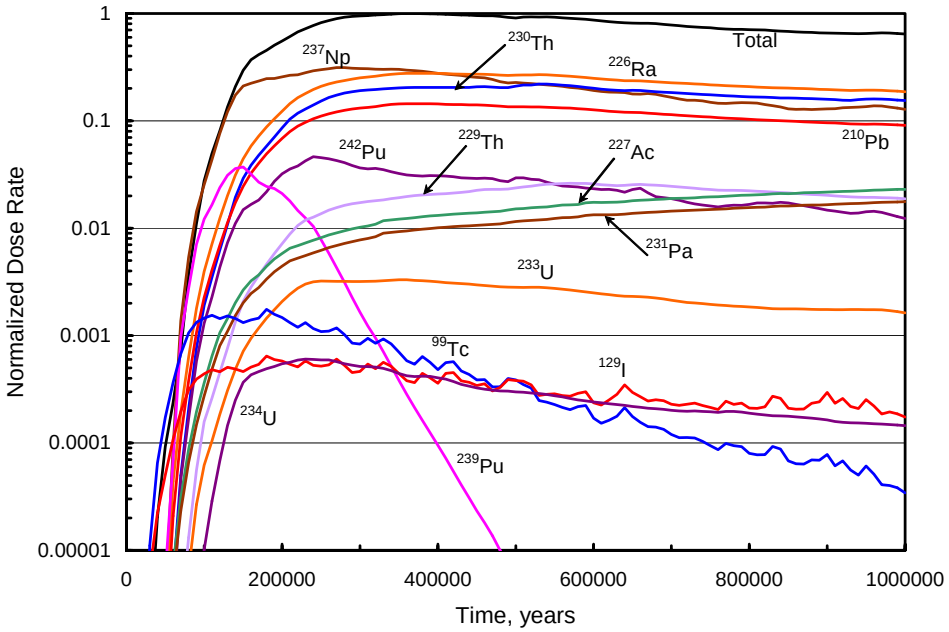

Fig. 1. Total and isotopic dose rates for direct disposal of PWR fuel as evaluated with the SSR model.

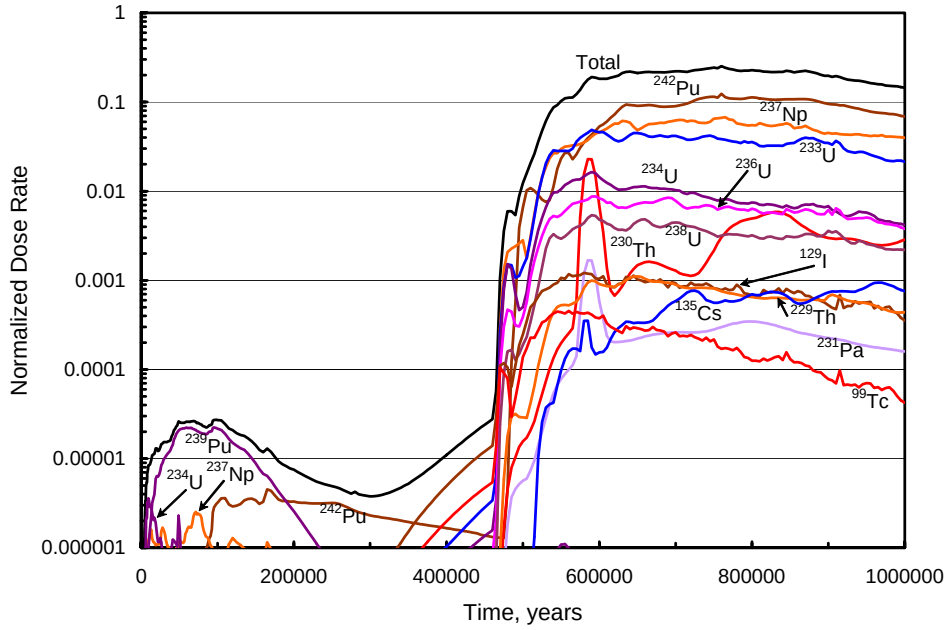

Fig. 2. Total and isotopic dose rates for direct disposal of PWR fuel as evaluated with the US model.

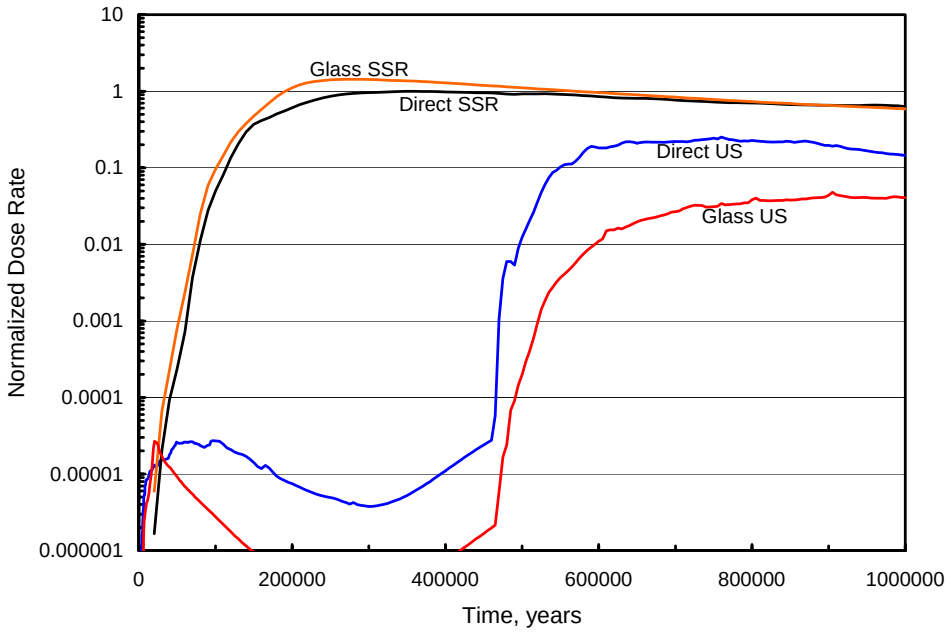

Fig. 3 Comparison of SSR and US calculations of total dose rate for PWR fuel for directly disposed and glass waste forms.

Nuclear Engineering Division
Argonne National Laboratory
9700 South Cass Avenue, Bldg. 208
Argonne, IL 60439-4842

www.anl.gov

UChicago ▶ Argonne LLC

A U.S. Department of Energy laboratory
managed by UChicago Argonne, LLC